

**IN THE UNITED STATES PATENTS AND
TRADEMARKS OFFICE**

APPLICANT: Martin Melchiors Et Al.
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FOR: Aqueous Dispersions
Art Unit: 1711
Examiner: R. A. SERGENT

DECLARATION

I, Martin Melchiors, residing at Leichlingen, D-42799, Germany, declare as follows:

- 1) that I have studied chemistry at the University of Aachen from 1987- 1992;
- 2) that I received the degree of doctor rer. nat. at the University of Aachen in the year of 1995;
- 3) that since 1995 I am employed by Bayer AG, D-51368 Leverkusen, Germany;
- 4) that I am working in the research field of aqueous coating compositions since 1998 and that I am familiar with the subject matter of the above identified application;
- 5) that the following tests were carried out under my supervision and control:

I) Blocked polyisocyanate (according to the invention, see example B1)

1250 g of Desmodur® N 3300 (aliphatic polyisocyanate based on hexamethylene di-isocyanate, Bayer AG), 208 g of 1-methoxypropylacetate-2 and 418 g of Solvent-naphtha 100 (Shell) were heated to 50°C. 628 g of 3,5-dimethylpyrazole were added in such a way, with stirring, that the temperature did not exceed 65°C. Stirring was then continued at 50°C until no more isocyanate could be detected by IR spectroscopy.

II) Aqueous dispersions of polyol containing urethane groups and blocked polyisocyanate (according to the invention, see example D9).

2478 g of a polyester with an OH content of 5.4% and an acid value of 3 mg KOH/g containing 2.0% of propylene glycol, 35.0% of neopentyl glycol, 10.4% of trimethylolpropane, 30.0% of isophthalic acid and 22.6% of adipic acid were charged to a 4 l reaction vessel with a cooling, heating and stirring device and heated to 130°C together with 150 g of dimethylolpropionic acid, 125 g of N-methylpyrrolidone and 3.8 g of tin octoate and homogenised for 30 min. The mixture was then cooled to 100°C and 372 g of hexamethylene diisocyanate (HDI) were added with vigorous stirring, the mixture was heated to 130°C (using the exothermic nature of the reaction) and kept at this temperature until no more NCO groups could be detected.

The resin melt then had a viscosity of 121 seconds, determined as the efflux time of a 50% xylene solution in a DIN 4 cup at 23 °C.

336 g of the blocked polyisocyanate B1 were then added to 900 g of this resin melt at 65°C, the mixture was homogenised for 20 min at this temperature and 28.7 g of dimethylethanolamine were added. Stirring was continued for 10 min at 65°C and then 1230 g of dist. water were added slowly.

The dispersion obtained had a solids content of 43.8%, a co-solvent content of 4.7%, a viscosity of 840 mPas and an average particle size of 80 nm. The acid value was 19.6 mg KOH/g, the OH content 2.6% (in each case based on 100% solids content).

III) Urethane-modified polyester resin without blocked polyisocyanate

267 g (3.1 mol %) of 1.2 propylene glycol, 5080 g (43.6 mol %) of neopentyl glycol, 1419 g (9.5 mol %) of trimethylolpropane, 3093 g (18 mol %) of phthalic anhydride and 4101 g (25.1 mol %) of isophthalic acid were weighed out into a 15 liter reaction vessel equipped with stirrer, cooler, heater and water separator. The mixture was heated to 220°C and condensed with elimination of water until the acid number was 3.

8450 g of this polyester, 398 g of dimethylolpropionic acid and 100g of neopentyl glycol were dissolved in 780 g of N-methylpyrrolidone, 10 g of dibutyltin dilaurate were added as catalyst and the mixture was heated to 75°C. After addition of 1150 g of hexamethylene diisocyanate, the temperature was raised to 125°C with the heat generated by the exothermic reaction. Stirring was continued until no further NCO

groups were detectable in the reaction mixture. 268 g of DMEA were then added and the product was dispersed in 11.270 g of water.

An approximately 45 % solids dispersion was obtained having an organic solvent content of approximately 3.5 %. The content of organically bound hydroxyl groups was 1.5 % (based on the dispersion).

A) Preparation of clear coats

a) According to the invention

150.0 parts by wt. of the dispersion according to example II) were formulated to a waterborne clear coat with 1.2 parts by wt. of a commercial flow additive (Additol® XW 395, Vianova Resins), 1.2 parts by wt. of a commercial wetting agent (Surfynol® 104, 50 %), 3.0 parts by wt. of 10 % aqueous solution of NMP and 19.6 parts by wt. of dist. water, applied to glass panels (wet film thickness 120 µm), allowed to dry for 10 min at room temperature and then stoved for 30 min at 140 °C and 160°C. Table 1 shows the results of the paint tests.

b) Comparison example: not according to the invention

125.0 parts by wt. of the dispersion according to example III) were formulated to a waterborne clear coat with 25.0 parts by wt. of the blocked polyisocyanate I), 1.4 parts by wt. of a commercial flow additive (Additol® XW 395, Vianova Resins), 1.4 parts by wt. of a commercial wetting agent (Surfynol® 104, 50 %), 3.6 parts by wt. of 10 % aqueous solution of DMEA and 28.0 parts by wt. of dist. water, applied to glass panel (wet film thickness 120 µm), allowed to dry for 10 min at room temperature and then stoved for 30 min at 140 °C and 160°C. Table 1 shows the results of the paint tests.

Table 1:

Example	(a)	(b) (comparison)
Drying 10 min RT +30 min 140°C		
Appearance of paint film	Satisf.	Strongly structured surface, insufficient film formation
Pendulum hardness	74	not possible because of the surface defects
Solvent insistence	3344	not possible because of the surface defects
Drying 10 min RT +30 min. 160°C		
Appearance of paint film	Satisf.	Strongly structured surface, insufficient film formation

Pendulum hardness	80	not possible because of the surface defects
Solvent resistance	3244	not possible because of the surface defects

B) Waterborne stoving primer surfacers

Preparation of a pigment paste P

A predispersed slurry composed of 4.78 g of 70 % water-thinnable polyester resin (Bayhydrol® D 270, Bayer AG), 9.33 g of dist. water, 0.68 g of 10% dimethyl-ethanolamine in water and 0.61g of commercial wetting agent (Surfynol® 104, 50 %), 12.26 g of titanium dioxide (Tronox® R-FD-I, Kerr McGee Pigments GmbH & Co KG), 0.14 g of black iron oxide (Bayferrox® 303 T), 12.34 g of barium sulfate (Blanc fixe Micro, Sachtleben GmbH), 3.01 g of talc (Micro Talc IT Extra, Norwegian Talc) and 0.42 g of antisettling agent (Aerosil® R 972 Degussa) was ground to a paste for 30 minutes in a commercial pearl mill, with cooling.

a) Preparation of a waterborne stoving primer surfacer, according to the invention

144.2 parts by wt. of II) were ground to a paste with 132.5 parts by wt. of the pigment paste P described above, 8.6 parts by wt. of a commercial melamine resin (Maprenal® MF 904, Vianova Resins) and adjusted to a pH of 8.2 ± 0.5 with 30.0 parts by wt. of dist. water and 0-2 parts by wt. of a 10% aqueous solution of dimethylethanolamine and to a viscosity of 40 ± 5 seconds efflux time at 23°C in the DIN 5 cup.

These coatings were applied by means of a spray gun with a nozzle diameter of 1.5 mm and an atomisation pressure of 5 bar to glass panels in a resulting dry film thickness of 25 µm to 35 µm. The resulting wet coating films were allowed to dry at room temperature and then stoved in a circulating air oven for 25 min. at 165°C.

b) Comparison example, not according to the invention

90.9 parts by wt. of the dispersions according to III) were ground to a paste with 132.5 parts by wt. of the pigment paste P, 8.6 parts by wt. of a commercial melamine resin (Maprenal® MF 904, Vianova Resins) and 18.2 parts by wt. of polyisocyanate I) and adjusted to a pH of 8.3 ± 0.5 with 32 parts by wt. of dist. water and 0.5 parts by wt. of a 10% aqueous solution of dimethylethanolamine and to a viscosity of 40 ± 5 seconds efflux time at 23°C in the DIN 5 cup.

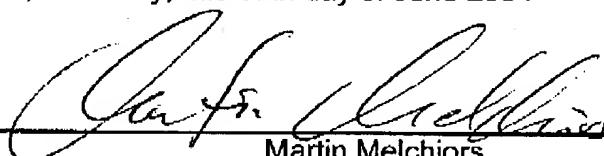
The surfacer was applied with a wet film thickness of 120 µm to glass panels, allowed to dry for 10 min at RT and then stoved for 25 min at 165°C. Table 2 shows the results of the paint tests.

Table 2:

Example	(a)	(b) (comparison)
Surfacer on glass (10 min RT +25 min 165°C		
Stability of liquid (ready-to-use) coating	Strong viscosity decrease after 10 d 40°C	stable
Gloss 20°/ 60°	49/86	21/69
Pendulum hardness	120 s	108 s
Appearance of film	satisf.	Structured surface, gel particles

I further declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Signed at Leverkusen, Germany, this 07th day of June 2004



Martin Melchior